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BASIC RESEARCH ON MIST FLAMMABILITY--PHASE I, EXPERIMENTAL FACILITY DEVELOPMENT

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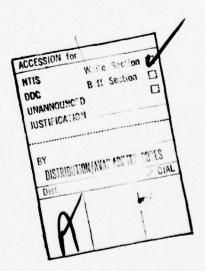
ARO 14298.1-CX AFLRL 197 ABSTRACT

9 Final rept. 1 Jun 76-31 Jul 77

A rugged, modular flame tube has been developed for use in studying laminar flame speeds and lean limits of both premixed vapors and heterogeneous mist systems. The flame tube design incorporates a mist inlet slit where optical measurements can be made. The thin sheet of mist then expands in the flow and fills the cross-section of the flame tube. flame tube comprises a series of 76 mm ID bolosilicate-glass "beaded process pipe" fittings as modules of the flame tube assembly. Each of the 254 mm long modules comprises a "reducer cross". The mist generator module, ignitor module, and sampling modules comprise the 76 mm ID "reducer cross" with 25 mm ID side ports; while the light-scattering module comprises a "reducer cross" with 51 mm ID side ports to be used as windows. Nylon plugs, machined to fit the side ports, provide means of access into the flame tube in each module. The 76 mm ID modules are joined by single-bolt couplings with polytetrafluoroethylene gaskets and hydrin liners designed for use with this process pipe. An air blast atomization nozzle is utilized for mist generation. This commercially-available nozzle which proved to operate satisfactorily, basically aspirates the liquid fuel into the air stream of an ultrasonic whistle. spark-gap assembly with a 25,000 volt power supply was supported in a nylon plug and inserted into the ignitor module. The remainder of the flame tube consisted of a series of sampling modules, each containing a 2 mm ID sample probe. The sample

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lines from the probes were electrically heated to insure vaporization. The withdrawn samples of fuel/air mixture were passed through a platinized asbestos/copper oxide wire catalyst bed maintained at about 400°C. The equivalence ratio of the mixture was then determined from the measured CO and CO2 formed by the essentially complete oxidation of the hydrocarbon. Forward light scattering equipment was assembled, but this was not used during this initial year of the study period. The system comprised a helium neon laser, that passes through the optically thin droplet/air sheet. The resulting scattered light is focused onto a detector with sensitive angular adjustment. The light scattering data can be rapidly reduced to a particle size distribution by means of a computer program. Observation and measurement of flame propagation was accomplished with the use of black-and-white video tape recordings of the experiments. Playback and single-frame evaluation provided information not only on flame position versus time but also on changes in the flame shape and thickness as it propagates through the tube. Two flame tube configurations are discussed. In the first attempt, the flame speeds were unusually high, ranging from 1 to 25 meters/second for vapors and from 1 to 75 meters/second This appeared to result from the onset of turbulent for mists. combustion. In the second attempt, a more conventional flame tube configuration was adopted. In this final flame tube configuration, the derived flame speeds and lean limits of gas phase heptane/air mixtures were in good agreement with literature values. The results obtained with liquid mists suggested that upward propagation would be most appropriate because of droplet sedimentation and wall-coalescence effects. However, no such experiments were conducted during this initial year of the study.



#### INTRODUCTION

The unwanted burning of hydrocarbon fuel represents a significant hazard to Army personnel and equipment, especially during combat. Fuel fire vulnerability problems experienced with rotary wing aircraft resulted in extensive Army-sponsored research on the development of fire-safety fuels. Prior to 1969, these efforts were concerned with stiff (semi-solid) aqueous JP-4 emulsions (and gels). (1) The emphasis then shifted to low-consistency (semi-liquid) aqueous JP-8 (and Jet A-1) emulsions. (2) However, these were eventually supplanted by dilute JP-8 solutions of extremely high molecular weight polymers that inhibit atomization (antimist fuels). (3,4)

Development of the helicopter "crashworthy fuel system" by the Army alleviated (for survivable helicopter accidents) the post-crash fire problem for rotary-wing aircraft. However, post-crash fires in fixed-wing aircraft and in-flight incendiary threats continue to represent a need for reduced fuel fire vulnerability. Hence, Army-sponsored research on the flammability and performance characteristics of antimist aircraft fuels has continued. In addition, the Army is presently conducting research on fire resistant fuels for use in ground combat (diesel) vehicles, and this involves the use of mistaltering techniques in addition to other flammability mitigation approaches.

<sup>(1)</sup> Weatherford, W.D., Jr. and Gray, J.T., "Modified Fuels for Improved Army Helicopter Safety", DOD/Rand "Second Symposium on Increased Survivability of Aircraft", February 1970.

<sup>(2)</sup> Weatherford, W.D., Jr. and Schaekel, F.W., "Emulsified Fuels and Aircraft Safety", AGARD/NATO 37th PEP Meeting "Aircraft Fuels, Lubricants, and Fire Safety", The Hague, Netherlands, May 1971 (AGARD-CP-84-71, pp. 21, 1-21, 12).

<sup>(3)</sup> LePera, M.E. and Weatherford, W.D., Jr., "U.S. Army Fuel Developments", "Industry-Military Jet Fuel Quality Symposium", January 1973.

<sup>(4)</sup> Weatherford, W.D., Jr. and Wright, B.R., "Status of Research on Antimist Aircraft Turbine Engine Fuels in the United States", AGARD/NATO 45th PEP Meeting "Aircraft Fire Safety", Rome, Italy, April 1975.

It has been well established that fuel mist, when present, plays an important role in flame initiation and propagation, even with volatile fuels, such as gasoline and military jet fuel  $(JP-4)^{(5)}$ , as well as with low-volatility fuels, such as aviation kerosine, and diesel fuel<sup>(6)</sup>. The physical and chemical processes involved in such phenomena are among the least studied and understood of all combustion phenomena.

The authors have initiated a basic research study, sponsored by the Army Research Office, to clarify ambiguous (or anomalous) published information on mist flammability. The multiyear program, as originally proposed, comprises quiescent (or nonturbulent) mist flammability studies that attempt to recognize and characterize influences of the range and distribution of droplet diameters, physical properties, chemical composition, and space-time history. It is the purpose of this manuscript to provide an interim (first-year) review on the status of the program, including descriptions of the experimental equipment developed for this study.

<sup>(5)</sup> Pinkel, I.I. et al, "Mechanism of Start and Development of Aircraft Crash Fires", NACA Report No. 1133, 1953.

<sup>(6)</sup> Pinkel, I.I. et al, "Origin and Prevention of Crash Fires in Turbojet Aircraft", NACA Report No. 1363, 1958.

### TECHNICAL BACKGROUND INFORMATION

## Mist Flammability

The following background discussion defines the major differences between flame initiation and propagation in, (1) heterogeneous mixtures of liquid droplets with air and, (2) homogeneous mixtures of fuel vapor with air, and it offers technical explanations for such differences. The purpose of the basic research to be conducted with the equipment and techniques described in this manuscript will be to provide quantitative information about such effects and to develop an improved understanding of the mechanisms of such heterogeneous combustion.

The flammability limits of mists are strongly influenced by the droplet size distribution of the dispersed liquid phase. In fact, on the basis of hydrocarbon weight per unit air volume, the lower flammability limit for a mist can be less than that of the same hydrocarbon in a homogeneous vapor-air mixture. It is partly because of this latter phenomenon that fuel mists represent a significant flammability hazard even though their ignition energy is greater than that of gaseous fuels.

This anomaly in the lower limit of flammability stems partly from the fact that, at the surface of each drop, the fuel vapor concentration varies from the equilibrium vapor pressure through a decreasing concentration range to zero (or near zero) in the bulk air surrounding the drop. Hence, if the equilibrium vapor pressure prior to ignition, or the dynamic vapor pressure during exposure to thermal effects of burning neighboring droplets, exceeds the lower limit of flammability, a flammable mixture may occur somewhere between the drop surface and the bulk air phase, irrespective of the overall equivalence ratio. In such cases, flames can propagate from drop to drop

unless the mist dispersion is so dilute that the thermal effects of burning drops cannot achieve ignition of neighboring drops.

In some cases, the anomalously low lean flammability limits of mists have been attributed to fuel droplet sedimentationenrichment of the flame front. (7) In these cases, such sedimentation effects were believed to increase the flame-front fuel concentration to nearly the same value as would be required for the lean limit in a homogeneous system. However, more recent research has indicated that liquid droplet suspensions with drop sizes exceeding 10 µm display flamefront lean limits that decrease significantly as the drop size increases. (8) This phenomenon is attributed to different mechanisms of flame spread in heterogeneous, relative to homogeneous, systems. Even if the sedimentation mechanism were responsible for "apparent" low lean limits, such limits could be applicable to vehicle crash or other fuel mist producing situations. In such cases, the flame front could propagate perpendicularly or obliquely to sedimentation generated fuel droplet concentration gradients, and a little bit of fuel could go a long way toward generating a flammable mist.

Flame propagation rates through fuel mists may be faster than in vapor phase fuel/air mixtures, the extend depending upon the characteristics of the particular mist dispersion and the base fuel composition. This enhanced flame velocity of mists has been attributed by some to the presence of acetylenes and hydrogen formed by pyrolysis in the "thicker" flame. (7) However, this has not been confirmed as the sole mechanism.

<sup>(7)</sup> Burgoyne, J.H., "Mist and Spray Explosions", Chem. Eng. Progress, 53, 121M-124M (1957).

<sup>(8)</sup> Burgoyne, J.H., "The Flammability of Mists and Sprays", "Proc. Sec. Symp. Chem. Proc. Hazards, Inst. Chem. Engrs." (London), 1963 (also, Fire Res. Abs. and Rev., 9, No. 2, 101 (1967).

Flame-speed enhancement could also result from the increased "expansion factor" (increase in the number of moles in the vapor phase due to the passage of the flame front) that results when liquid fuel droplets are enveloped by the flame front and subsequently vaporized and burned. Flame-speed enhancement could also stem from the entirely different propagation mechanism observed with large droplets (relative to homogeneous systems). In the light of these unique properties of flammable mists, the importance of such phenomena relative to fuel fire safety should not be underestimated. In fact, it is because of these properties that so much research and development attention has been (and is currently being) devoted to preventing or inhibiting fuel mist formation during ballistic or crash-impact events.

### Literature Search

Upon initiation of this study, a collection of selected pertinent references was assembled to document the "state of the art" in various facets of the study. The results are presented as an annotated bibliography in Appendix A. It is emphasized that this is not intended to represent a comprehensive bibliography, but rather, was assembled to provide a working summary of selected pertinent references.

#### EXPERIMENTAL PROGRAM

Examination of appropriate literature, such as that detailed in the Appendix, established that measurement of mist droplet diameters of less than about 0.5  $\mu m$  as part of this program would not be feasible. Moreover, the short optical path lengths required (to avoid multiple, successive scattering) with forward light scattering techniques precluded the use of a constant-diameter cylindrical mist confinement system. The latter requirement stems from the following considerations.

For typical hydrocarbon fuels, it is well known that the concentration of fuel in air at the lean flammability limit is about 50 mg/l. At this concentration of fuel dispersed as liquid droplets, the optical path length, that causes a 10% eclipse of the cross-sectional are the droplet-air mixture by droplets can be calculated. Fo a 10% eclipse, it is believed that multiple, successive might scattering would not cause significant errors in calculated droplet size distributions. (9) tuitously, this optical path length, expressed in mm, is approximately numerically equal to the eclipsing droplet diameter, expressed in µm. Obviously, higher or lower liquid fuel concentrations would result in respectively lower or higher optical path length requirements for the 10% eclipse. For the 10-100 μm mean diameter droplets anticipated for this study, the optical path length would have to be as short as one cm to satisfy the above-described criterion. On the other hand, good experimental practice requires that the surface-to-volume ratio of the mist confinement system must be as low as practical in order to minimize surface influences on flammability characteristics. Accordingly, the flame tube design selected for this study incorporates a mist inlet slit where optical measurements

<sup>(9)</sup> Personal communication with Dr. R.E. Beissner of Southwest Research Institute.

can be made through the entering thin sheet of mist before the sheet enlarges to eventually fill the cross-section of the flame tube. Details of the flame tube design, droplet size measurement methodology, gas sample analysis technique, and flame propagation and observation procedures are presented in the following description of the experimental studies.

## Modular Transparent Flame Tube

Consideration was given to various types of equipment that could be used for flame speed measurement. Several desired criteria precluded the use of laboratory glassware or opaque pipe or tubing. These criteria were:

- 1. Nonfragile construction,
- 2. Transparent windows or walls, and
- 3. Off-the-shelf availability of the major components.

All of these criteria were met by the selection of borosilicateglass "beaded process pipe" fittings\* as the modules for the flame tube assembly. The tube size selected for this assembly was 76 mm ID (3-inch) with a wall thickness of about 5 mm (7/32-inch). Each of the modules comprises a 254 mm long (10inch) "reducer cross" with 25 mm ID (1-inch) side ports and 76 mm ID end openings. (As will be noted later, the reducer cross utilized for the mist entry slit and light-scattering viewing windows comprises the basic 76 mm ID tube with 51 mm ID (2-inch) side ports.) Each of the 76 mm ID modules is attached to the adjacent module(s) with a single-bolt coupling designed for use with this process pipe. The beaded ends of the pipe are cushioned within each coupling in polytetrofluorethylene (PTFE) gaskets with hydrin liners. Nylon plugs were machined to a size that nearly fills each of the side ports of the reducer crosses, and entry holes were drilled through these

<sup>\*</sup>Kimax "beaded process pipe", manufactured by Kimble Glass Co., Inc.

nylon plugs as required for each module. The outer end of each nylon plug is in the form of a bead matching that of the glass side port and was secured with a single-bolt process pipe coupling matching the size of the particular side opening. The nylon-filled, double-side-port geometry was selected so that a multiplicity of penetrations into the flame tube can be made, as desired. Each of the modules comprising the flame tube assembly is described in the following paragraphs.

Mist Generator Module - After preliminary experimentation with a sonic air blast atomization nozzle on hand,\* this nozzle was selected for use in the flame tube assembly. This nozzle is commercially available and is rated for unthrottled fuel rates up to 200 milliliters per minute induced by air rates up to 34 liters per minute. It is reported to produce mists having mean diameters as low as 20  $\mu m$ . Figure 1 illustrates the atomizing module wherein this nozzle is supported by 1/8-inch stainless steel tubes entering through the previously described nylon plug. As shown in Figure 2, a stainless steel micrometer needle valve is attached to the liquid fuel inlet line. In use, the desired fuel-air ratio is established by adjusting both the atomizing air pressure and the liquid fuel needle valve setting.

Light Scattering Module - The flame tube module designed for mist particle size measurement is illustrated in Figure 3. As noted previously, it comprises a "reducer cross" having 51 mm ID (2-inch) side ports. An 0-ring sealed, 10 mm thick partition supports a centrally located 50 mm OD by 25 mm long stainless steel cylinder (1.6 mm wall). The downstream end of this cylinder is closed with a flat stainless steel disc (1.6 mm thick) containing a 1 mm by 40 mm slit oriented perpendicular to the axis of the 51 mm ID side ports. It is intended to install a window disc in each of the 51 mm ID side ports

<sup>\*</sup>Sonicore Model No. 052H, manufactured by Sonic Combustion Systems, Inc.

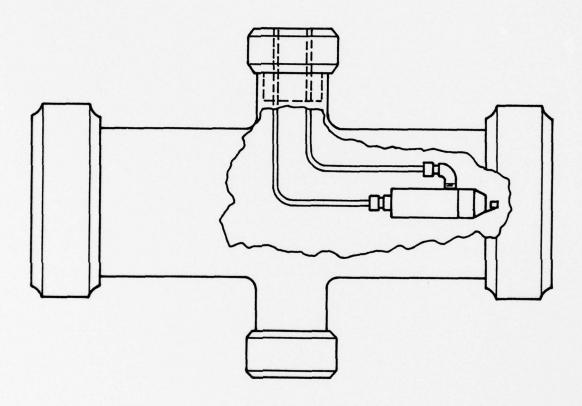


FIGURE 1. SONIC AIRBLAST ATOMIZATION NOZZLE IN FLAME TUBE MODULE .

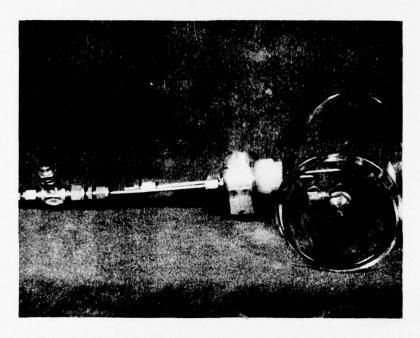


FIGURE 2. PHOTOGRAPH OF SONIC AIRBLAST ATOMIZATION NOZZLE IN FLAME TUBE MODULE

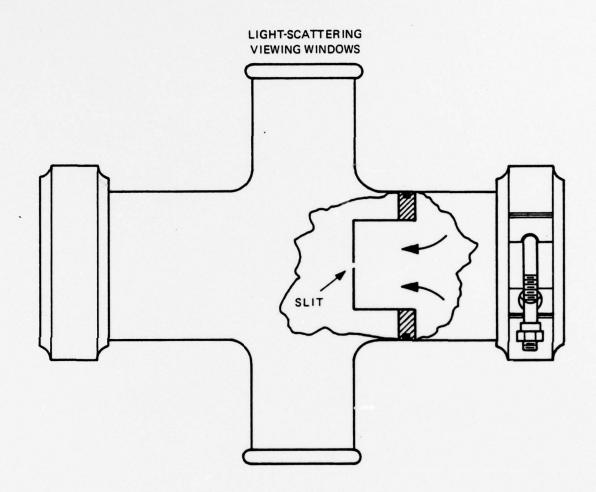


FIGURE 3. MIST ENTRY SLIT IN FLAME TUBE MODULE

when light scattering measurements are to be made. The window fixtures will be so designed that a small bleed air stream will flush mist away from the window to prevent fogging. Because mist droplet size measurements were not conducted during this first year of the program, these window fixtures were not installed; rather, temporary opaque closures (rubber stoppers) were inserted.

Ignitor Module - A spark gap assembly was inserted through one of the previously-described nylon plugs in one of the 76 mm ID by 25 mm ID "reducer crosses" as illustrated in Figure 4. The electrodes (3 mm diameter with a 3 mm gap) are made of stainless steel, and they are energized by a 25,000 volt power supply.

Sampling Modules - Figure 5 illustrates the sampling system presently employed for determining the fuel/air stoichiometry. A straight, blunt-end 3 mm OD (1/8-inch) stainless steel tube (2 mm ID) is inserted through one of the nylon plugs in the 76 mm ID by 25 mm ID "reducer cross". The module is oriented so that the flow through the probe leading to the catalytic oxidation unit is downward. A toggle valve is provided in this line directly adjacent to the nylon plug, and the remaining 3 mm OD tubing leading to the catalytic oxidation unit is wrapped with flexible electric heating tape. A thermocouple placed beneath this tape, and the heater voltage is adjusted to maintain an observed temperature of at least 135°C (275°F).

# Sample Analysis System

Under the conditions anticipated for mist flammability studies, the equivalence ratio will always be less than unity. Hence, there will always be sufficient excess air present to convert all of the fuel carbon content into carbon dioxide. Accordingly, a catalytic oxidation technique was selected for measuring the overall equivalence ratio of the probe samples.

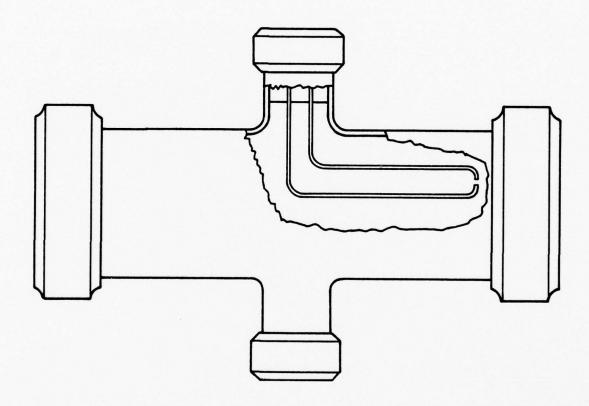


FIGURE 4. SPARK GAP IN FLAME TUBE MODULE

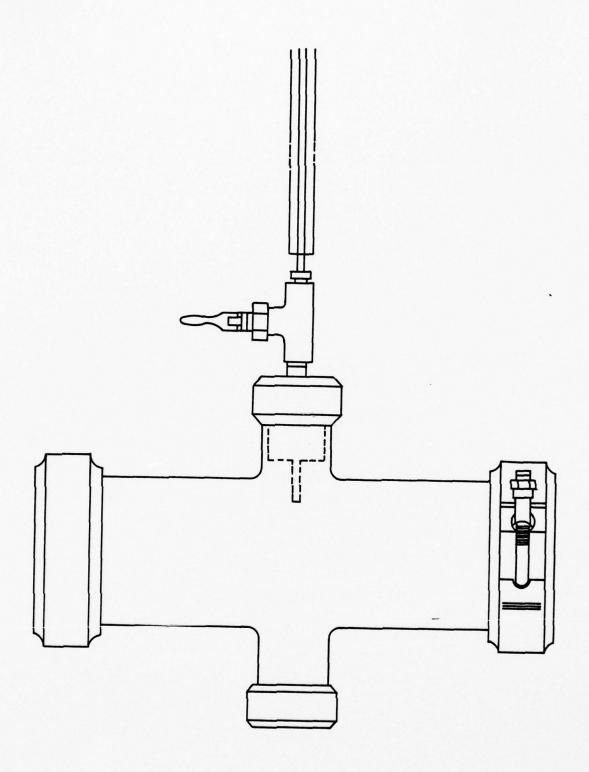


FIGURE 5. SAMPLE WITHDRAWAL FLAME TUBE MODULE

The catalytic oxidation unit developed for this purpose is illustrated in Figure 6. It comprises a stainless steel block containing an imbedded cartridge heater and thermocouple and a cavity packed with a mixture of platinized asbestos and copper oxide wire. The primary purpose of the copper oxide wire is to provide a loose matrix to prevent excessive pressure drop through the catalyst bed.

Brief experimental studies were conducted to confirm satisfactory operation of the catalytic oxidation unit. Ambient temperature air was bubbled through n-heptane liquid, and the effluent air, saturated with n-heptane, was passed through the catalytic unit, maintained at about 400°C (750°F), to the carbon dioxide measurement system. The mixture of platinized asbestos and copper oxide wire was found to be effective at 350-400°C for producing the theoretical carbon dioxide content corresponding to the complete oxidation of the n-heptane in the lean fuel/air mixture.

# Droplet Size Measurement Technique

Upon initiation of this program, a detailed review was conducted of published information on techniques for observing and documenting droplet or particle sizes in liquid mists and aerosols. Among those methods that were claimed to be applicable to particles of less than 0.5  $\mu$ m diameter, a combination, three-way particle size measurement technique proposed in the literature (10) for particle diameters ranging from 0.01 to 100  $\mu$ m was judged unsuitable (11) for this study.

<sup>(10)</sup> Shifrin, K.S., and Perelman, A.Y., "Inversion of Light Scattering Data for the Determination of Spherical Particle Spectrum", "Proc. Sec. Interdisciplinary Conf. on Electromagnetic Scattering, Univ. of Mass., 1965", Rowell, R.L., and Stein, R.S., ed., Gordon and Breach, New York, 1967, pp. 131-167.

<sup>(11)</sup> Review and/or recommendation by Dr. R.E. Beissner of the Southwest Research Institute staff.

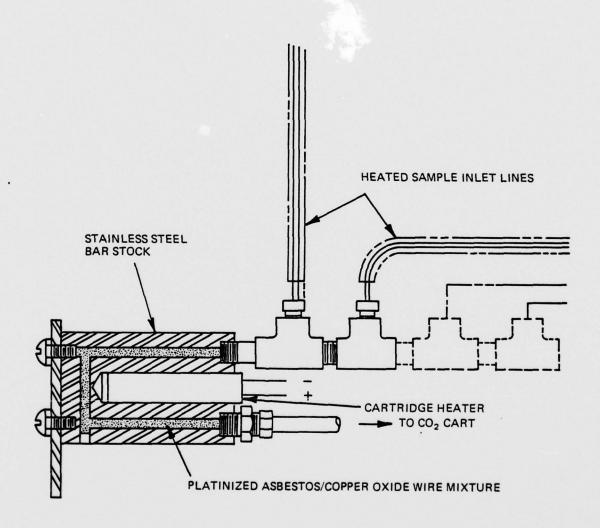


FIGURE 6. CATALYTIC OXIDATION UNIT FOR EQUIVALENCE RATIO MEASUREMENT

Also, a combination sedimentation-light scattering technique  $^{(11,12)}$  was also considered as an optical approach for this study because it is theoretically applicable to particles smaller than the wavelength of the light being scattered. However, serious difficulties are reported for particles of less than 0.5  $\mu$ m diameter because of convection-origin perturbations of sedimentation rates. A holographic technique  $^{(13)}$  would be suitable for in situ observation of non-burning droplet size distributions for droplets larger than about 1.0  $\mu$ m. Such a technique would entail substantial data-reduction effort, and the required level of effort and expenditures were considered beyond the scope of this study.

The droplet size measurement technique selected for use in this study is basically the same as that originally proposed. The forward light scattering method described in Appendix B is accomplished by passing an expanded collimated helium-neon laser beam through an optically thin droplet/air zone. scattered light is focused onto a detector by a condensing lens (focal length approximately 12 inches). All optical components on the optical bench are adjustable. (photo-diode) mount has a micrometer adjustment and a dial indicator to measure scattered light intensities at precise displacements from the center line (incident laser beam). The size distribution including the mean droplet diameter and sauter mean diameter is computed from the measured light intensities with a program written by Watkins and Opdyke of AVCO/ Lycoming. The program breaks the distribution up into size groups which can be fitted to a Rosen-Ramler distribution function for sprays.

Sample problems have been worked out using light scattering data obtained in this laboratory from a fan tip spray nozzle

<sup>(12)</sup> Kerker, M., "The Scattering of Light", Academic Press, New York (1969), pp. 388-396.

<sup>(13)</sup> Such as that studied by Polymeropolous, et al (See Appendix A).

and a Hewlett-Packard 9820 calculator for which the program was written. All of the light-scattering system components were procured by SwRI, and the hardware was adapted for omnidirectional utilization. Unfortunately, however, the mist flammability work did not reach the state of development during this first year where it would have been meaningful to make droplet size distribution measurements.

## Observation and Measurement of Flame Propagation

Although the "cross" geometry of the individual modules of the flame tube would permit insertion of flame detectors (e.g., fiber optics, photodiodes, ion probes, etc.), it was deemed more appropriate in the present study to record the entire visible experiment on video tape. Playback and single-frame evaluation provides information not only on flame position versus time, but also on changes in the flame shape and thickness as it propagates through the tube. In fact, the selection of the "glass pipe" flame tube design stemmed in part from this laboratory's video tape capabilities.

The time resolution of the video tape is 1/60th of a second per half frame, and the flame position versus time could be established by counting half-frames. However, this tedious data reduction procedure was circumvented by utilizing the output of an electronic clock superimposed upon the taped video image. This electronic clock provided a recorded time resolution of 1/100th of a second; hence, the flame position versus time could be thereby established with a time resolution of the same order of magnitude as the video half-frame period. Incidentally, the first four digits of the clock read-out were intended by the manufacturer to have represented the month and year, but these digits were utilized in this study to designate the identity of the fuel and the number of experiments conducted with that fuel. Thereby, each tape recording fully identified the experiment and documented the results.

## Operation of Modular Flame Tube

Exploratory Studies - The previously described flame tube modules were assembled in a horizontal flame propagation mode. An exploratory configuration of the module assembly (flame tube) is shown in Figure 7 along with the sample processing accessories. The various modules were arranged in this configuration in the following order, from bottom to top of the photograph in Figure 7 (in the direction of air flow): mist generator (directed downstream), ignitor (with spark gap at downstream end of module), light scattering module (with mist entry slit fixture removed and windows closed with stoppers), and four successive sampling modules (with only the first two connected to the catalytic oxidation unit). A vacuum pump was connected (via a pebble-filled knock-out drum, ball valve, and flame arrestor) to the downstream end of the flame tube. The inlet end of the mist generator module was covered with a 16 mm thick plate containing a centrally located 25 mm diameter hole. A cork stopper was lightly rested in this hole to prevent air from being drawn into the apparatus through this hole. This stopper was subsequently blown out (with negligible resistance) when ignition occurred. One of the side ports in the last sampling module was kept open to allow some forward flow of unburned gas ahead of the advancing flame.

For premixed vapor experiments, such as that shown in Figure 8, the fuel vapor was introduced through the liquid port of the atomizing nozzle, and air was introduced through the air inlet of the nozzle. A mist ignition experiment is shown in Figure 9 (note the mist fog in Figure 9a prior to ignition). In these experiments, the fuel was aspirated into the atomizing nozzle through a micrometer needle valve, and the relative fuel and air rates were varied by adjusting both the air pressure and the needle valve setting. In the case of either prevaporized fuel or liquid mist, the resulting stoichiometry was monitored by observing the CO<sub>2</sub> content of the catalyst unit effluent,

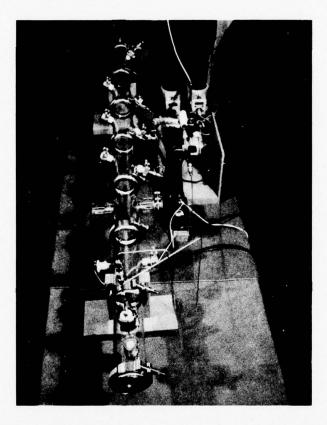
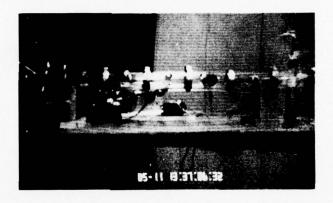


FIGURE 7. PHOTOGRAPH OF MODULAR FLAME TUBE (EXPLORATORY CONFIGURATION) AND SAMPLE PROCESSING ACCESSORIES



a. Room Lights on Without Ignition

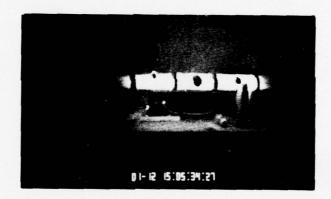


b. 0.12 Seconds After Ignition

FIGURE 8. VIDEO TAPE STILL FRAMES OF n-BUTANE FLAME  $(\phi$  = 0.99) ( EXPLORATORY CONFIGURATION)



a. Room Light on Without Ingition



b. 0.04 Seconds After Ignition

FIGURE 9. VIDEO TAPE STILL FRAMES OF JP-5 MIST FLAME  $(\phi = 0.06)$  (EXPLORATORY CONFIGURATION)

and the outlet end of the flame tube was closed with the ball valve prior to attempting ignition.

The preliminary experiments conducted with this exploratory configuration of the flame tube strikingly demonstrated that the configuration was not optimum. Figure 8 illustrates a typical flame observed with a stoichiometric n-butane/air mixture. The observed flame speed for this fuel/air system varied from greater than 25 to less than 1.3 meters per second. The simultaneous illumination of three consecutive modules shown in Figure 9 for a JP-5 mist/air mixture represents an observed flame speed possibly as high as 75 meters per second, while in other similar experiments, flame speeds of less than 1.3 meters per second were observed.

The laminar burning velocity (S,) of a gas phase stoichiometric hydrocarbon/air mixture is about 40 cm per second. The observed laminar flame speed (S<sub>c</sub>) depends strongly on the configuration of the flame speed measuring device, but it seldom exceeds a value of 2 to 3 meters/second. An upper-limit laminar flame speed of 2 to 3 meters/second is achieved when the burned gas behind the flame is confined and the thermal expansion (approximately 7:1) develops a burned gas speed ( $S_{\sigma}$ ) that must be added to the burning velocity. Furthermore, the observed flame speed  $(S_s)$  in a tube is higher than  $S_{11}$  because the actual area of the flame front (semi-ellipsoidal due to poiseulle flow) is approximately a factor of 2 larger than the cross-sectional area of the flame tube. Hence, the burning velocity in a tube must be expressed as a/A  $(S_s - S_g)$  where a/A is the area ratio of the tube to the flame sheath. The extreme range and the anomalously high values of  $S_s$  observed using the flame tube configuration described above appears to be due to the onset of turbulent combustion. Depending on the magnitude of slight fluid disturbances in the ignition and initial flame propagation events, it is conceivable that the turbulent intensity and rate of growth of turbulence could vary widely.

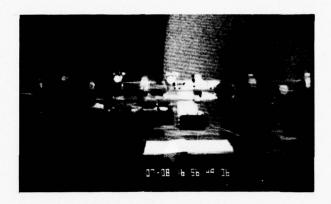
these experiments, the ignition occurred midway (actually closer to one end) in the flame tube, and openings of about one inch in diameter were placed at each end of the tube to prevent a pressure buildup. Because the flame was propagating in both directions with the hot burned gas expanding in between, it was estimated that the observed flame speed should be in the 2 to 3 meter/second range. A calculation of the Renolds number, assuming a slug of gas moving at a velocity of 2 meters/second in a 76 mm (3-inch) diameter tube, gives a value of about 10,000. This is well above the basic criteria (Renolds number > 2300) for turbulence. This may also account for several cases where it appeared as if the flame was accelerating and cases where it would propagate at a near normal rate (approximately 2 meters/ second) and then suddenly accelerate rapidly. It seems evident that these observations were due to turbulent growth in the flame duct.

# Final Flame Tube Configuration

To resolve this difficulty in measuring a laminar flame speed, a more conventional configuration was used in which one end of the tube was left completely open and a relatively small hole (7 mm diameter) was placed at the other end. The ignition source was set a few centimeters inside the tube at the open end. In testing the new system, premixed heptane/air mixtures of varying equivalence ratios were bled into the flame tube and flame speeds were measured with the video tape instrumentation (see Figure 10). The observed flame speeds, as shown in Figure 11, are in the 70 to 100 cm/second range, depending on equivalence ratio. The inflammability limit equivalence ratio for the heptane/air mixture in this particular flame tube was 0.7; a slightly high value that may have resulted from some air dilution at the open end of the tube. However, this value of 0.7 falls near the range of previously measured values. (14,15)

<sup>(14)</sup> Zabetakis, M.G., Bulletin 627, Bureau of Mines, "Flammability Characteristics of Combustible", 1965.

<sup>(15)</sup> McCrocken, D.J., "Hydrocarbon Combustion and Physical Properties", BRL Report No. 1496, September 1970.



a. Room Lights on Without Ignition



b. 0.071 Seconds After Ignition

FIGURE 10. VIDEO TAPE STILL FRAMES OF n-HEPTANE FLAME  $(\phi = 1.0)$  (FINAL CONFIGURATION)

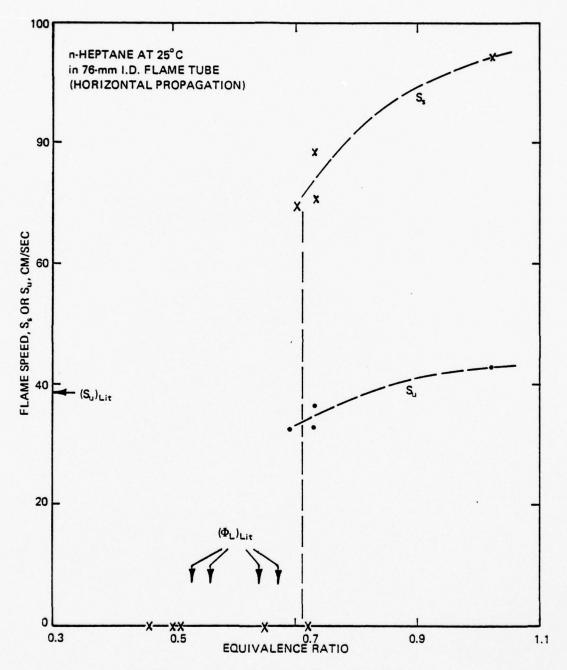


FIGURE 11. PRELIMINARY CALIBRATION OF MODULAR FLAME TUBE WITH  $n\text{-}\mathsf{HEPTANE}$ 

A simple, tentative model was developed to evaluate the burning velocity  $(S_u)$  from the observed flame speed  $S_s$ . It was assumed that there was negligible pressure buildup behind the flame due to the expanded hot burned gas since that end of the tube was wide open and gave no resistance to flow. What remained then was the pressure buildup by the passage of the flame front into the unburned gases, i.e.,

$$\Delta P = \rho_u S_u^2 \left[ \frac{\rho_u}{\rho_b} - 1 \right]$$

This differential pressure must be balanced by the pressure drop due to flow of unburned gas through the 7 mm diameter orifice at the tube end. This pressure balance may be expressed as

$$\rho_{\mathbf{u}} S_{\mathbf{u}}^{2} \left[ \frac{\rho_{\mathbf{u}}}{\rho_{\mathbf{b}}} - 1 \right] (A/a) = 32 L\mu G/\rho_{\mathbf{u}} D^{2}$$

where the mass flux through the orifice  $G = 118.5 \ \rho_u S_g$ ,  $\rho_u$  is the unburned gas density and  $\rho_b$  is the burned gas density,  $\mu$  is the viscosity, D is the orifice diameter (0.7 cm), L is the length of the orifice tube (26 mm) and A/a is the ratio of flame sheath area to the tube cross-sectional area (approximately 2). It is assumed that  $\rho_u/\rho_b$  is simply the ratio of the flame temperature, for a given stoichiometry, to the ambient temperature  $(T_b/T_u)$  multiplied by a small correction for the change in number of moles (n) and that  $S_u = a/A$   $(S_s-S_g)$ . The burning velocity may then be evaluated from the quadratic equation below.

2 
$$\left(n \frac{T_b}{T_u} - 1\right) S_u^2 + 6.2 \times 10^3 S_u - 3.1 \times 10^3 S_s = 0$$

The results of these calculations are shown in Figure 11 where a burning velocity of 43 cm/second is obtained at a near-stoichiometric condition. This appears to be a reasonable value of  $S_{\rm u}$  for heptane considering the scatter in measured values for the most well documented case, methane. An accepted

value for the flame speed of heptane at stoichiometric conditions is 38.6 cm/second. (15) Because the value of 43 cm/second is in reasonably good agreement with this value, it is concluded that the experimental method of measuring flame speed would be adequate for future work on mists and sprays.

Based upon mist settling and coalescence effects observed during mist ignition experiments with an exploratory flame tube configuration (Figure 7), it was apparent that the flame tube would have to be mounted in a vertical position for mist experiments. Provision would then be required for collecting and removing coalescenced liquid from the upstream side of the mist entry slit (Figure 3). Unfortunately, after the described final flame tube configuration was established and the preliminary, horizontal propagation calibration data (Figure 11) were obtained with n-heptane vapor, insufficient time remained in the initial year of the program to reorient and modify the flame tube in its final configuration for liquid mist studies. There may still be problems with droplet deposition on the flame tube walls even by placing the tube in a vertical arrangement, but these difficulties can only be resolved by further experimentation and improvement of the state of the art.

### CONCLUSIONS AND RECOMMENDATIONS

A rugged, modular flame tube comprising off-the-shelf components, for the most part, has been developed and shown to be effective for measuring flame speeds and lean flammability limits of homogeneous fuel/air mixtures. Exploratory studies with liquid JP-5 mists indicate that vertically upward propagation is most appropriate for such systems because of droplet sedimentation effects.

It is recommended that the study be continued, with emphasis on documenting flame speeds, lean limits, and droplet size distribution (greater than 1  $\mu m)$  for various multicomponent and single component liquid fuels and other flammable fluids in the Army inventory. The objective of such studies would be to develop sufficient understanding of mist flammability phenomena to allow development of analytical models to predict and correlate such phenomena.

#### ACKNOWLEDGMENTS

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Beissner and C.A. Moses, and Mr. S.A. Cerwin for developing the design of the forward light scattering technique for droplet size distribution measurement.

#### APPENDIX A

#### ANNOTATED BIBLIOGRAPHY OF SELECTED PERTINENT REFERENCES

### MIST AND SPRAY COMBUSTION

1. Gollahalli, S.R., "Buoyancy Effects on the Flame Structure in the Wakes of Burning Liquid Drops", Combustion and Flame, 29, 21, (1977).

The flame structure in the wakes of burning methanol and n-heptane droplets in a downward air flow is studied. Temperature profiles, composition profiles, radiant power emitted from the flame, extention velocity, flame length, are all examined and compared with earlier studies in an upward air flow. It was concluded that inverted flames (those in downward air flows) "are shorter, produce less soot and emit less radiation than upright flames" (flames in upward air flows).

Putnam, A.A., Benington, F., et al., "Injection and Combustion of Liquid Fuels", Wright Air Development Center Technical Report 56-344, Astia Document No. AD 118142, March 1957.

"This Monograph contains a review of material in the unclassified literature relating directly to the fundamental physical phenomena involved in steady flow processes in high-intensity combustors. To systematize the presentation, the report has been divided into six parts; as follows:

"Part I. Atomization of Liquid Fuels: Chapter 1. "The Mechanism of Atomization", Chapter 2. "Methods of Atomization", Chapter 3. "Design of Atomizers", and Chapter 4. "Spray Analysis".

<u>Part II.</u> <u>Ballistics of Droplets:</u> Chapter 5. "Ballistics of a Single Droplet" and Chapter 6. "Dynamics of Dispersions".

"Part III. Evaporation of Droplets: Chapter 7. "The Thermodynamics and Kinetics of Evaporation", Chapter 8. "Single-Droplet Evaporation", Chapter 9. "Evaporation of a Moving Droplet" and Chapter 10. "Spray Evaporation".

"Part IV. Fluid Dynamics: Chapter 11. "The Equations of Fluid Dynamics", Chapter 12. "Turbulence and Chapter 13. "Hydrodynamic Recirculation".

"Part V. Homogeneous Combustion: Chapter 14. "Laminar Flame Propagation", Chapter 15. "Turbulent Flames of Premixed Gases", Chapter 16. "The Stability Limits of

Premixed Flames" and Chapter 17. "Ignition of Combustible Mixtures".

"Part VI. Heterogeneous Combustion: Chapter 18. "Droplet Combustion" and Chapter 19. "Diffusion Flames".

3. Hedley, A.B., Nuruzzaman, A.S.M. and Martin, G.F., "Progress Review No. 62: Combustion of Single Droplets and Simplified Spray Systems", <u>Journal of the Institute of Fuel</u>, 38, 38, (1971).

"Over 200 papers concerning the combustion processes of atomized fuels are discussed under the main headings:
(i) single droplets, (ii) droplet arrays, (iii) monodisperse sprays, and (iv) polydisperse sprays. It is shown that quasi-steady state theories of single droplets cannot explain adequately the mechanism of combustion and that the recent experimental results emphasize the unsteady nature of combustion. It is concluded that much more information is required on the mechanics of mixing, droplet interference, flame stabilization, and such before a rigorous theoretical treatment of spray combustion can be attempted satisfactorily" (225 references).

4. Spalding, D.B., "Some Fundamentals of Combustion", Butterworths Scientific Publications, London, 1955.

pp. 122-143: This reference is concerned with the heat and mass transfer taking place with the chemical reaction when liquid fuels are used. "The transfer number of envelope flames, the combustion of fuel droplets, the droplets in motion, the transient effects, the drag, the wake of flames, and the combustion of fuel sprays" are discussed in some detail.

pp. 217-222: This reference is concerned with combustion without premixing. Topics discussed include "flames stabilized by recirculation with incomplete premixing, flames in uniform suspension, partially mixed flames, and partially mixed flames with interphase mass transfer.

5. Beer, J.M. and Chigier, N.A., "Combustion Aerodynamics", Applied Science Publishers Ltd., London, 1972.

pp. 147-195: In Chapter 6 of the above reference, "Droplets and Sprays" are discussed. Included in this chapter are the following subjects: Atomization of Liquid Fuels; Drop Formation in Sprays; Interaction of Air Streams and Sprays; Combustion of Sprays; Single, Droplet Combustion; The Combustion of Droplet Arrays, Streams and Monodisperse Sprays; Twin Fluid Atomized Liquid Fuel Diffusion Flames; and Pressure Jet Oil Flames.

pp. 247-260: In Chapter 8 of the above reference, "Measurements in Flame", some of the subjects discussed are:

Measurements of droplets sizes, velocities and angles of flight using high speed photography; and turbulence measurements in flames.

6. "Liquid-in-Gas Dispersions", Chemical Engineers' Handbook, Fifth Edition, Perry, R.H. and Chilton, C.H., editors, McGraw-Hill Book Company, New York, 1973, Chapter 18, pp. 58-68.

This reference discusses the three types of liquid-ingas dispersion that are of interest to those in the process industries. These are "sprays produced by nozzles and other atomizing systems for combustion, mass/heat transfer, or coating of surfaces; entrainment generated by gas bubbling through a liquid as in a distillation tower or kettle reboiler; fogs generated from gases that are supersatured". The break-up process for these three are discussed under the topics: liquid-column break-up, liquid-sheet break-up, droplet break-up, spray nozzles, pressure nozzles, two-fluid atomizers, droplet-size distribution and fog condensation.

7. "Particle Dynamics", "Chemical Engineers' Handbook", Fifth Edition, Perry, R.H. and Chilton, C.H., editors, McGraw-Hill Book Company, New York, 1973, Chapter 5, pp. 61-65.

This reference discusses the particle dynamics of spherical rigid particles, non-spherical rigid particles, gas bubbles, liquid drops in liquids and liquid drops in gases. It contains a table for drag coefficient and related functions for spherical particles.

8. Williams, A., "Combustion of Droplets of Liquid Fuels: A Review", Combustion & Flame, 21, 1, (1973).

"A review is presented of recent developments of the combustion of single droplets with the object of stimulating discussion and future work in the field. Among the areas covered are the combustion of stationary and moving droplets in an oxidizing atmosphere, the combustion of monopropellants, the influence of high pressures and the ignition of droplets. The application of droplet theories to the modelling of various combustion systems is also outlined" (216 references).

9. Longwell, J.P., "Combustion of Liquid Fuels, High Speed Aerodynamics & Jet Propulsion", Vol. II, Section J, "Combustion Processes", Lewis, B., Pease, R.N. and Taylor, H.S., editors, Princeton University Press, Princeton, NJ, 1956.

Topics discussed are atomization, including the air blast nozzle used by Nukiyama & Tanasawa, the pressure-atomizing nozzles, such as studied by Longwell (Chem. Eng. Sc. D.

thesis, Mass. Inst. Technol., 1943) and a similar study of Joyce (Lloyd, P. Power Jets Special Rpt. 510). The mixing of fuel sprays by turbulent diffusion is described along with a brief discussion on the precipitation of fuel sprays. Another topic discussed is the evaporation of fuel drops. Equations for calculating the equilibrium vaporization are given and a method for calculating the evaporation rate is described. Also included in this reference is a discussion of the combustion of fuel drops.

10. Law, C.K., "Unsteady Droplet Combustion with Droplet Heating", Combustion & Flame, 26, 17 (1976).

"Unsteady droplet combustion caused by droplet heating is modelled by assuming quasi-steady gas-phase processes and the droplet temperature being spatially uniform but temporarlly varying. Results show droplet heating is a significant source for the experimentally observed unsteady combustion phenomena of Okajima and Kumagai."

11. Law, C.K., "Multicomponent Droplet Combustion with Rapid Internal Mixing", Combustion & Flame, 26, 219 (1976).

"Two models are proposed to describe the gas-phase diffusioncontrolled, unsteady combustion of a multicomponent droplet in a stagnant, unbounded atmosphere. The first, termed the Ideal-Mixture Model, assumes that the mixture behaves as an ideal mixture in its phase change characteristics, and that the composition and temperature within the droplet are spatially uniform but temporarily varying. Expressions are obtained for the droplet vaporization rate and other quantities of interest. Sample solutions indicate that the components vaporize approximately sequentially in the order of their relative volatilities, and that the vaporization rate is insensitive to the mixture composition during combustion as well as during pure vaporization in hot environments. Available experimental evidence supports the theoretical model. The second model, termed the Shell Model, assumes a shelled distribution of the components such that quasi-steady, single-component vaporization prevails for each shell. Simplified solutions are derived and are shown to closely approximate the bulk vaporization behavior described by the more detailed Ideal-Mixture Model, particularly for the prediction of the total vaporization time".

12. Natarajan, R., "A Shadowgraphic Investigation of High Pressure Droplet Vaporization", Combustion & Flame, 26, 407 (1976).

This paper describes the investigation of high pressure droplet vaporization using cold n-pentane drops that are "injected into helium or nitrogen pressures up to  $4250 \, \mathrm{kN/m^2}$  and temperature up to approximately  $533 \mathrm{K}$ ."

13. Radusch, R., "On the Vaporization of Water Drops", Chemie-Ingenieur-Technik, 28, 275 (1956).

The author uses the geometric variations that take place during the evaporation of a droplet and the heat transfer associated with it, along with several empirical relationships and derives conclusions about droplet vaporization. Most of these conclusions are confirmed in the laboratory.

14. Williams, F.A., "Combustion Theory", Addison-Wesley Publishing Co., Inc., Reading, Massachusetts (1965).

pp. 256-259: In this section of the book (Chapter 11) equations are given in order to obtain the droplet size distribution. Equations for some of the spray properties are also given.

pp. 274-287: This section of chapter 11 describes the heterogeneous laminar flame, the overall continuity and the spray equation for it. The author discusses the species conservation, the momentum, energy and state. The mathematical problem and boundary condition are outlined and some simplications that can be made under certain conditions. Included in this section is a comparison of theory and experimental results of the heterogeneous burning velocity of a monodisperse fuel spray.

15. Williams, F.A., "Spray Combustion Theory", Combustion and Flame, 3, 226-228 (1959).

The coupled steady-state equations of motion of a statistical ensemble of spherical droplets and a coexisting multicomponent reacting gaseous continuum are derived under the assumption that the induced random fluctuations in the properties of the fluid are negligible. These equations are reduced to a form appropriate for describing the one-dimensional flow of a fluid containing a spray which is stationary with respect to the gas. The burning-rate eigenvalue problem for one-dimensional heterogeneous combustion processes of the type  $A \rightarrow nB$  is formulated, and an analysis for sprays composed of uniform droplets illustrates the properties of the eigenvalue. An approximate analytical solution for the burning velocity is shown to be in numerical agreement with experiments on tetralin.

Mizutani, Y. and Nakajima, A., "Combustion of Fuel Vapor-Drop-Air Systems: Part I. Open Burner Flames", Combustion and Flame, 24, 343 (1973).

"The burning characteristics of fuel vapor-drop-air systems (propane-kerosene drop-air systems) have been examined using an inverted-cone-flame-burner apparatus.

A small amount of kerosene drops added to a propane-air mixture markedly accelerates the burning velocity for an overall fuel-air ratio and extends the region of stable burning leanwards. A small amount of propane added to a kerosene drop-air mixture, however, never exhibits such combustion-promoting effects. There exists an optimum value in the quantity of kerosene drops to be added. The weaker the intensity of turbulence in the flame zone, or the finer the drops, the greater combustion-promoting effects are observed. Kerosene mists consisting of submicron droplets, however, exhibit no such combustion-promoting effects."

17. Onuma, Y. and Ogasawara, M., "Studies on the Structure of a Spray Combustion Flame", Fifteenth Symposium (International) on Combustion, 1974, pp. 453-465.

"To clarify the flame structure of a spray burner, the following experiments and analysis were carried out. (1) Droplet and temperature distributions, flow velocity, and gas composition were measured in the flame of an airatomizing burner. It was found that the region where the droplets exist is limited to a small area above the burner nozzle. From the correlation between the above various distributions, it was concluded that most of the droplets in the flame do not burn individually, but that fuel vapor from the droplets concentrates and burns like a gas diffusion flame. (2) Various measurements were then made on a spray combustion flame and a turbulent gas diffusion flame under the same conditions. Comparing the two sets of data, it was found that the flames are similar in structure. (3) Assuming that the droplets evaporate in the flame, their behavior was analyzed by making use of the knowledge which has been obtained for a single droplet. The calculated results were in fairly close agreement with the experimental results.

The above facts suggest the possibility that the spray combustion flame could be treated theoretically by applying the information for a single droplet and for a turbulent gas diffusion flame."

18. Waldman, C.H., "Theory of Non-Steady State Droplet Combustion", Fifteenth Symposium (International) on Combustion, 1974, pp. 429-442.

"The method of matched asymptotic expansions is applied to the analysis of non-steady state combustion of liquid fuel droplets. Asymptotic solutions are derived for inner and outer regions representing the droplet vicinity and far field, respectively. The expansions are based on a small parameter identified as the ratio of the droplet radius to the diffusion field radius. The outer region is dominated by unsteady diffusion; convection is

a higher-order effect. Viewed from the outer region, the droplet appears to be a point source of mass and sink for heat. The inner region is in a quasi-steady state and is characterized by a balance between convection and diffusion. The mass burning rate is governed by the inner-region solution; to lowest order the quasi-steady value is recovered. It is shown that perturbations due to the unsteady outer region increase the burning rate and reduce the droplet lifetime. Flame trajectories calculated from uniformly valid composite expansions show that (i) the flame moves away from the droplet then back towards it, (ii) the ratio of flame-to-droplet radius increases continually, and (iii) flame extinction occurs after the droplet vanishes. These results contradict the quasi-steady state theory of droplet combustion, but they agree with observed droplet behavior. Theoretical predictions exhibit good agreement with experimental data."

19. Spalding, D.B., "Combustion of a Single Droplet and of a Fuel Spray", Selected Combustion Problems, AGARD Combustion Colloquium, Butterworths Scientific Publications, 1954.

"The physical and chemical aspects of liquid fuel burning are shown to be capable of separate treatment. Considerable progress has been made in understanding the former aspect, and the burning rate of an isolated droplet can be predicted with acceptable precision if the natures of the fuel and atmosphere and of their relative motion are specified. The relative motion within a spray is, however, too complex to permit analysis as yet, and it is not even certain whether droplet burning or jet mixing processes control the flame length in gas-turbine combustion chambers. Knowledge of the conditions for the stable burning of liquid fuels is still less advanced, and is restricted to the laminar diffusion flame adjacent to a fuel surface. It appears that the extinction condition is related to the laminar flow speed but not to the spontaneous ignition characteristics of the fuel. contribution to design of the fundamental knowledge obtained so far has been largely negative in character. More direct application of this knowledge is only likely if the design of combustion systems is so modified as to be amenable to analysis. In the present state of the art this means restricting the use of fuel sprays."

20. Faeth, S.M., "Current Status of Droplet and Liquid Combustion", presented at Central States of the Combustion Institute, March 1977.

"The present understanding of spray combustion in rocket engine, gas turbine, diesel engine and industrial furnace applications is reviewed. In some cases, spray combustion can be modeled by ignoring the details of spray evaporation and treating the system in the same manner as a gaseous diffusion flame; however, in many circumstances, this type of simplification is not adequate and the turbulent twophase flow must be considered. The behavior of individual droplets is a necessary component of two-phase models and recent work on transient droplet evaporation, ignition and combustion is considered, along with a discussion of important simplifying assumptions involved with modeling these processes. Methods of modeling spray evaporation and combustion processes are also discussed including: onedimensional models for rocket engine and prevaporized combustion systems, lumped zone models (utilizing wellstirred reactor and plug flow regions) for gas turbine and furnace systems, locally homogeneous turbulent models, and two-phase models. The review highlights the need for improved injector characterization methods, more information of droplet transport characteristics in turbulent flow and continued development of more complete two-phase turbulent models."

21. Chigier, N.A., "The Atomization and Burning of Liquid Fuel Sprays", Prog. Energy Combus. Sci., 2, 97 (1976).

A review covering "high speed photography of droplets in sprays", "air blast atomization", "idealized spray flame", "physical model of spray combustion in a gas turbine engine", "air blast spray flames", "pressure jet, swirl atomized hollow cone spray flame", "single droplet combustion", "vaporization of droplets in gasair systems" and "atomization and evaporation of fuel sprays in diesel engines" (43 references).

22. Chigier, N.A., "Instrumental Techniques for Studying Heterogeneous Combustion", presented at the Central States Section of the Combustion Institute, March 1977.

"Diagnosis of heterogeneous combustion systems requires the measurement of physical and chemical properties in flames laden with liquid and solid particles. For liquidfueled combustors, the characteristics of the spray require to be determined, including the trajectories of individual droplets and their interaction with gas streams. Vaporization rates can be determined from measurement of the rate of change of diameter of individual droplets. Particulate matter emitted from combustion chambers is mainly in the solid phase and consists of both unburned fuel and non-burnable material. With heavy fuel oils, particles of the order of 100 µm can be emitted while, for the lighter fuels, particulate emissions are mainly in the sub-micron size region. The laser anemometer can be used for simultaneous measurement of velocity and particle size of individual droplets and particles in spray flames. Laser diffraction techniques can be used for

measurement of over-all size distributions. Laser Raman spectroscopy offers the possibility of simultaneous measurement of temperature and species concentration in flames. Presence of particles results in laser irradiated particulate heating and incandescence, which can lead to the swamping of signals. The use of coherent anti-Stokes Raman and near-resonant Raman offers possibilities for making laser Raman measurements in particle laden flames.

Solid probes for measuring temperature and species concentration require special adaptation when used in particle laden flows. Particles may damage probes by direct impingement and cause blockage of orifices. Thermocouples can be used in sprays, but the effects of direct droplet impingement must be considered. Diagnostic techniques have been developed in which thermocouple signals are digitized and recorded on a computer. This system permits the rapid measurement of time constants and, subsequently, digital frequency compensation of temperature time histories, providing a frequency response of the order of 1 kHz. Information on spray boundaries is provided by both laser anemometer and thermocouple measurements. Gas sampling probes, specially designed to separate liquid and solid particles from the gas, are used for removal of samples from within the flame. Size analysis of particles is made by micro-photography and computer image analyser. Species concentration in the gas is measured by gas phase chromatography, flame ionization detectors, non-dispersive infra-red and ultra-violet detectors and chemiluminescent analysers.

All measurement diagnostic techniques are affected by the presence of large-scale coherent eddy structures in the turbulent flow systems. The need for increasing frequency response, recording and analysis of variations with time and the use of conditional sampling in intermittent flows is emphasized. The coupling of high-speed movies with laser optical probes to give simultaneous recording of visual and signal events is recommended."

23. Gerstein, M. and Graves, C.C., "Some Aspects of Combustion of Liquid Fuel", "Combustion Researches and Reviews, 6th and 7th AGARD Combustion Panel Meetings", Butterworths Scientific Publications, London, 1955.

The paper discusses diffusion flames and these factors associated with them. Topics include "Burning of Fuel Sprays in Air Streams", "Fuel Spray Spreading", and "Fuel Spray Burning in Turbojet Combustors" (25 references).

24. Berlad, A.L. and Killory, J., "Flame Propagation and Extinction for Clouds of Particles", presented at the Central States Section Meeting, The Combustion Institute, March 1977.

The paper is concerned with flame propagation and extinction theory and experiment for uniformly premixed clouds of (vaporizable) particulates in a gravity-free environment.

25. Hayashi, S. and Sehchiro Kumagi, "Flame Structure of Droplet-Vapor-Air Mixtures", Archimum termodynamiki i Spalania, 6, 479 (1975).

"The structure of flames in laminar droplet-vapor-air flows has been studied for several fuels with a wide range of volatility. The droplet-size distribution is relatively narrow; the mean and maximum droplet diameters are about 25 and 40  $\mu m$ , respectively, for every test.

In terms of the flame structure, three different types have been observed depending on the vapor concentration in the mixture: (a) burning of individual droplets in their own flames; (b) burning of droplets surrounded by local flames behind a continuous premixed flame; and (c) a continuous flame front followed by droplets evaporating in the burned gas.

The effects of the size and number density of droplets on the flame structure have also been discussed."

### DROPLET SIZE DISTRIBUTION

# Light Scattering Methods:

26. Webster, J.M., Weight, R.P., and Archenhold, E., "Holographic Size Analysis of Burning Sprays", Combustion & Flame, 27, 395 (1976).

This paper describes the technique of holography as applied to analyses of burning sprays. It permits the entire cloud of droplets within the expanded laser beam to be recorded. The authors describe the advantages and the disadvantages of this technique. Some of the advantages are "Its application does not distrub the flame or combustion process. A size analysis of droplets and their distribution in three dimensions can be obtained in a single record."

27. Dieck, R.H. and Roberts, R.L., "The Determination of the Sauter Mean Droplet Diameter in Fuel Nozzle Sprays", Applied Optics, 9, 2007 (1970).

"A technique has been developed for determining the Sauter mean droplet diameter in a nozzle spray by direct analysis of the small angle, forward scattered light profile. The spatial intensity of the profile was scanned with a microphotometer and the information recorded on punched paper tape for later reduction by computer. A knowledge of a particular functional form for the intensity profile

was required for computer processing of the data, and it was therefore determined empirically that the natural log of the intensity of the small angle, forward scattered light had a gaussian profile with respect to the scattering half-angle."

28. Kerker, M., Matijevic, E., et al, "Aerosol Studies by Light Scattering. I. Particle Size Distribution by Polarization Ratio Method", <u>Journal of Colloid Science</u>, 19, 213 (1964).

"The size distributions of very small aerosol particles  $(0.5 < \bar{\alpha} < 1.9)$  were determined by means of polarization ratio and/or dissymmetry factor measurements. The values of the polarization ratio and dissymmetry factor were calculated for various size distributions assuming a logarithmic normal distribution. The experiments were performed with linoleic acid, triphenylphosphate (TPP), and stearic acid aerosols, and the estimated values of geometric mean size and standard deviation were confirmed with the values obtained from electronmicroscopic observations."

29. Roberts, J.H. and Webb, M.J., "Measurement of Droplet Size for Wide Range Particle Distribution", AIAA Journal, 2, 583 (1964).

The author describes an extension of his earlier work where he developed "a method for the determination of a mean droplet diameter of a spray from the analyses of the small-forward-angle diffractively scattered light intensity." This was only applicable to what was described as an upper-limit distribution function (ULDF). This type was largely small droplets, like those used in spray studies. Thus this paper describes his investigation into the possible use of the diffractive-scattering measuring technique to distribution forms other than ULDF.

30. Takahashi, K. and Iwai, S., "Estimation of Size Distribution of Small Aerosol Particles by Light-Scattering Measurement", Journal of Colloid & Interface Science, 23, 113 (1967).

"The size distributions of very small aerosol particles (.05 <  $\bar{\alpha}$  < 1.9) were determined by means of polarization ratio and/or dissymmetry factor measurements. The values of the polarization ratio and dissymmetry factor were calculated for various size distributions assuming a logarithmic normal distribution. The experiments were performed with linoleic acid, triphenylphosphate (TPP), and stearic acid aerosols, and the estimated values of geometric mean size and standard deviation were confirmed with the values obtained from electronmicroscopic observations."

31. Dobbins, R.A., "Further Studies on the Light Scattering Technique for Determination of Size Distributions in Burning Sprays II. Wide Range Photographic Photometry", Aeronautical Engineering Laboratory Report No. 498, Air Research and Development Command, Contract AF 18(600)-1527 AFOSR TN 60-353 (1960).

"This report deals with the development and feasibility of a photographic technique for the measurement of dropletsize distribution in burning or evaporating fuel streams with an accuracy equal to, or greater than any presently known electronic measurement techniques.

The technique is based upon the relationship between angular distribution of scattered light and size distribution of the scattering particles."

32. Durbin, E.J., "Optical Methods Involving Light Scattering for Measuring Size and Concentration of Condensation Particles in Supercooled Hypersonic Flow", National Advisory Committee for Aeronautics, Technical Note 2441 (1951).

"The purpose of this paper is to provide experimental methods for measuring the size and concentration of condensation particles in supercooled hypersonic flow in order to help resolve some of the questions concerning the nature of this condensation."

"Optical methods involving light scattering for measuring the size and concentration of condensation particles in supercooled hypersonic air flow are discussed. Two methods based on scattered-light measurements and on transmitted-light measurements are given which can be used for obtaining quantitative measurements provided (1) that steady-state conditions can be achieved during the time required for the measurement of light intensity, (2) that the condensation particles are approximately spherical in shape and essentially uniform in size, and (3) that the index of refraction of the condensation particles is known approximately."

33. McVey, J.B., Kennedy, J.B., et al, "Diagnostic Techniques for Measurements in Burning Sprays", presented at 1976 Fall Meeting of the Western States Section of The Combustion Institute, October 1976.

Laser holography, laser velocimetry, and a phase-discriminating sampling probe have been used to characterize the spray produced by a pressure-atomizing fuel nozzle in a combusting flow. The instrumentation techniques employed are documented, and information obtained with the use of No. 2 distillate fuel is presented.

34. Penner, S.S., Bernard, J.M., and Jerskey, T., "Power Spectra Observed in Laser Scattering from Moving Polydisperse Particle Systems in Flames - I Theory, II Preliminary Experiments", Acta Astronautica, 3, 69-91, 93-105 (1976).

"Theoretical expressions are derived for the photocurrent power spectra observed with laser scattering from moving, polydisperse particle systems. The analysis refers to self-beating of scattered radiation and beating between the laser beams scattered in the same direction from two, equally-intense incident laser beams which arrive at symmetrical angles with respect to the scattering direction. When using a spherically-symmetric Gaussian distribution for the illumination intensity in the coherence volume (scattering region), a Voigt profile is obtained for the photocurrent power spectrum in the general case. The author has used the theoretical relations derived for moving systems in order to estimate the maximum values of the particle diameters which are measurable for Stokes-Einstein diffusion when radiation is scattered from He-Ne or Ar-ion lasers."

35. Anderson, W.L. and Beissner, R.E., "Counting and Classifying Small Objects by Far-Field Light Scattering", Applied Optics, 10, 1503 (1971).

"An approach to small particle counting and classification is proposed in which the size and shape distribution is determined from the far-field pattern of light scattered by the particles. By presuming that the scattering function for each particle species is known from auxiliary experiments or calculations, a general result applicable to irregular particles as well as simply shaped particles is obtained. Inversion formulas, by which the particle distribution is determined from the far-field data, are presented in forms suitable for digital or optical processing. An experimental approach to the design of a real-time optical counting and clssifying instrument is suggested."

36. Polymeropoulos, C.E., Sernas V., et al, "Generation of Polydisperse Sprays for Combustion Studies", presented at Eastern Section Meeting of The Combustion Institute, November 1975.

The paper describes the measuring of the droplet size distribution in kerosene-air sprays by means of the "far field" holographic technique. The droplet size distribution were measured for two sprays under controlled air flow and air-fuel ratio. The authors present the results by means of cumulative volume and number distributions of the sprays and these are correlated by using the upper limit log-normal probability distribu-

tion. The sprays were found to be suitable for combustion studies in that they appeared spatially uniform in their size distribution.

37. Polymeropoulos, C.E. and Sernas, V., "Measurement of Droplet Size and Fuel/Air-Ratio in Sprays", Combustion and Flame (1977).

"A holographic technique was employed to measure in situ the droplet size distribution and the fuel-air mass ratio in a flowing stream of kerosene spray. The halographic measurement of the fuel-to-air ratio was in good agreement with that obtained by direct fuel and air flow rate measurements. The upper limit log-probability distribution function provided a satisfactory correlation of the droplet size data.

38. Dobbins, R.A., Crocco, L., and Classman, I., "Measurement of Mean Particle Sizes of Sprays from Diffractively Scattered Light", AIAA Journal, 1, 1882 (1963).

"The angular distribution of scattering for polydispersions of particles distributed according to the Upper Limit Distribution Function is examined and is found to lack the sensitivity necessary to permit determination of size distribution. However, the volume-to-surface mean diameter is found to be directly dependent upon angular distribution of intensity for a wide variety of shapes of the distribution function. Therefore, the combination of both a scattering experiment together with a transmission experiment can be used to obtain both particle concentration and volume-to-surface mean diameter of particles in a spray. While there is no limitation with regard to the maximum diameter, the actual upper size limit that is measurable experimentally is controlled by considerations related to angular resolving power. Experimental results that show agreement between the volume-to-surface mean diameter as determined by scattering experiment and by microscopic examination are given for solid spheres."

39. Kerker, M., "Some Recent Reflections on Light Scattering", Journal of Colloid and Interface Science, 58, 100 (1977).

"Three recently discovered unusual light scattering effects are described in a qualitative way. These effects are concerned with: (1) nearly invisible particles, (2) internal heating of particles by radiation, and (3) Raman and fluorescent scattering by molecules embedded in particles."

40. Swithenbank, J., Beer, J.M., et al, "A Laser Diagnostic for the Measurement of Droplet and Particle Size Distribution", Report No. HIC 245, Department of Chemical Engineering and Fuel Technology, University of Sheffield.

"This study describes a technique which has been developed for the measurement of droplet or solid particle size distribution. ... The technique is based on the Fraunhofer diffraction of a parallel beam of mono-chromatic light by the moving droplets. A Fourier transform lens is used to focus a stationary light pattern onto a multi-element photo-detector to measure the diffracted light energy distribution. A mini-computer program translates the light energy distribution into the corresponding, unique, droplet size distribution. The droplets or particles are classified into 31 size groups spanning two decades of diameter, (e.g., 5  $\mu m$  to 500  $\mu m$  using a 300 mm focal length Fourier transform lens). Although the size range may be varied simply by changing the focal length of the lens, there is a lower limit of about 1  $\mu m$  for He/Ne laser light. ... "

41. Wang, C.P., "Laser Anemometry", American Scientist, 65, 291 (1977).

The paper describes a "powerful experimental technique" that uses light scattering and the Doppler's principle to measure velocity components.

## Other Methods and Combinations of Methods:

42. Beer, J.M., and Chigier, N.A., "Combustion Aerodynamics", Applied Science Publishers Ltd., London, 1972, pp. 246-253.

In surveying all the various techniques for measuring the drop size, the only one found to be suitable was high speed photography. This technique has been applied to the photography of an isothermal pressure jet spray with the air flow field uniform. It is now being used to include twin fluid atomizer flames, isothermal oil sprays and burning oil sprays. The latter two being stabilized by bluff bodies. The article shows a schematic of the experimental system used and describes its operation. A schematic of the spark units is also included.

43. Browning, J.A., "Production and Measurement of Single Drops, Sprays and Solid Suspensions", Advances in Chemistry Series, 20, 136 (1958).

"This review represents an excellent survey of the literature (284 references), principally of experimental work on the production and size measurements of single drops and of sprays. The emphasis is primarily on applications to combustion devices, although valuable information on sprays for fire righting is included. The following topics are discussed: Airstream Atomization (7 refs.); Pressure Atomization (27 refs.); Production of Aerosols (13 refs.); Single Drops and Streams of Uniform Drops (27 refs.); Spinning Disk Atomization (9 refs.); Solid Suspensions (36 refs.); Aggregation of Particles (10 refs.); Diffraction Theory and Equipment (44 refs.); Size Distribution

(9 refs.); Elutriation (4 refs.); Freezing of Drops (2 refs.); Jet Impactors (12 refs.); Photographic Measurements (12 refs.); Sedimentation (8 refs.); Wax Method (2 refs.)."

44. Benson, S.M., El-Wakel, M.M., et al, "Fluorescent Technique for Determining the Cross-Sectional Drop Size Distributions of Liquid Sprays", ARS Journal, May 1966, 447.

The development of a unique laboratory instrument which measures cross-sectional drop sizes in dynamic liquid sprays by a fluorescent technique is described. This technique consists of focusing a short duration, high intensity radiation flash into a thin sheet of radiation which intersects the spray nearly perpendicularly to the spray axis. The spray drops within this sheet, acting as primary radiators due to a fluorescent dye additive, are photographed and subsequently measured. The error in the measurement of the drop size was estimated from actual camera calibration data through the use of a specially made reticle to be less than 10 percent for 10-micron diameter drops, and was found to decrease sharply for larger diameter drops. The fluorescent, optical and photographic problems encountered in the development are discussed. Drop size distribution data and spray photographs showing drop sizes are presented.

45. Hirleman, E.D., Jr. and Wittig, S.L.K., "In Situ Optical Measurement of Automobile Exhaust Gas Particulate Size Distributions: Regular Fuel and Methanol Mixtures", Sixteenth Symposium (International) on Combustion, The Combustion Institute, Pittsburgh, PA, 1976.

Optical techniques provide in situ, real time analysis of particulate size distributions in raw exhaust gases thereby eliminating complications inherent in conventional sampling probe - inertial impactor or other interfering methods. A new Multiple Ratio Single Particle Counter (MRSPC) was designed for the in situ analysis of combustion generated particulates in the diameter range of 0.4 μm to 3.0 μm. Detailed calibration and testing confirmed the ability of the prototype to perform the desired measurements. The instrument was initially applied to the study of particulate size distributions emitted from a four cylinder Otto engine using regular fuel and methanol mixtures. The comparison with available data taken under different experimental conditions reveals considerable discrepancies and indicates the importance of engine system parameters as well as probe characteristics. Large increases in particulate emission rates were observed during acceleration tests confirming the observation of previous studies.

46. Simmons, H.C. and Lapera, D.J., "A High-Speed Spray Analyzer for Gas Turbine Fuel Nozzles", presented at the ASME Gas Turbine Conference, March 1969.

"This paper describes an instrument system designed to examine the spray characteristics of fuel nozzles, including drop size distribution, at specific positions in the spray cloud as well as the statistics relative to the entire spray. Additionally the spray mass density can be explored at different locations and the spray external shape accurately defined. The system uses large numbers of samples observed at high speed by a TV camera and the data is directly computerized."

47. Sundukov, I.N. and Cekalin, E.K., "Some Methods for Studying Two-Phase Fuel-Air Mixtures", Eighth Symposium (International) on Combustion, The Combustion Institute, 1962, pp. 1120-1125.

"A complex of methods is suggested for measuring the basic parameters of a two-phase polydispersed mixture in a free fuel-air jet. Measurements are made of the amount of the liquid phase of a readily evaporating fuel, the average size of the drops and the mean concentration of the fuel drops in a unit flow volume.

The above magnitudes are analyzed in their functional relationship."

## FLAMMABILITY LIMITS

48. Whittle, J., "Effect of Vaporization Rate on the Weak Combustion Limit of Liquid Sprays", <u>Fuel</u>, <u>XXXIII</u>, 192 (1954).

"Tests to determine the weak combustion limit have been carried out in an aero-type gas turbine combustion chamber on a number of fuels covering a wide range of volatilities. Two levels of fuel spray atomization were used. It has been found that the effect of changing from fine to relatively coarse atomization depends upon the volatility of the fuel. Directly opposite trends were obtained at the two extreme limits of volatility. An explanation of these results is given."

49. Roth, W. and Scheer, M.D., "Explosion Limit Phenomena and Their Use in Elucidation of Reaction Mechanisms", Advances in Chemistry Series, No. 20, 85 (1958).

The experimental observation of pressure limits for gaseous explosions and methods for determining explosion limits are discussed and critically evaluated. This includes the classical thermal theory of explosion-limit phenomena first mentioned by van't Hoff, as well as the modern branching chain hypothesis of Semenoff. The effect upon the explosion-limit pressures of temperature, composition, vessel diameter, and vessel surface

is considered for both thermal and branching-chain explosions. A treatment of the time factor (induction period) for the development of an explosion in quiescent gases in a closed system is also given. Several specific explosion reactions which have appeared in the literature within the past two or three decades are given in some detail. Emphasis is placed upon the use of explosion limit observations as a method for yielding information about the reaction mechanism.

50. Egerton, A.C., "Limits of Inflammability", Fourth Symposium (International) on Combustion, The Combustion Institute, 1952, pp. 4-13.

This is a survey on the subject from the time that Davy's first determined the limits of combustion of fire damp and air to the time of the symposium (1952). The standard procedure established by Gurgoyne and Williams-Leir is described, and their inflammability diagnosis for hydrogen, methane, ethylene, carbon monoxide, n-hexane, cyclohexane and benzene are discussed. Except for the last two compounds, where methyl bromide was used as the inert vapor in air, nitrogen was used as the inert gas in establishing the diagrams. These studies were extended by E.H. Coleman to include other halogenated compounds. Some of the other subjects covered in the review are: "the effect of additive on upper limit mixtures of carbon monoxide, of hydrogen, and of mixtures of these gases", "mixtures of air with mists of inflammable liquids and mixtures of air with dispersed solids", "cool flames", and "chain branching reactions".

51. Von Elbe, G., "The Problem of Ignition", Fourth Symposium (International) on Combustion, The Combustion Institute, 1952, pp. 13-20.

This survey covers selected material on ignition, as related to the formation of a combustion wave in an explosive medium. It reviews work related to the ignition of an explosive gas mixture by an electrical spark. Ignition by other sources, such as an electrically heated wire in an explosive medium, by heated spherical pellets moving through gas-air mixtures at various velocities, are also discussed.

52. Lovachev, L.A., "The Theory of Limits on Flame Propagation in Gases", Combustion and Flame, 17, 275 (1971).

The mechanism of flame extinction induced by convection is examined by the author. In order to obtain the flame propagation limits, a set of nonstationary equations for energy conservation, for diffusion of all reaction components, the Navier-Stokes equation, and the equation of continuity must be solved. The proposed mechanism can then be studied using this simplified model.

53. Diamy, A.M. and Ben-Aim, R.I., "The Explosion Limits of  $H_2$ - $O_2$ - $N_2$  Mixtures", Combustion and Flame, 15, 207 (1970).

A static system was used and the same procedure was used for determining the three limits. It was difficult for the authors to obtain accurate and reproduceable results for determining the limits. They found: it is difficult to say precisely whether the first limit determined is a flame limit or a slow-reaction limit; the second limit is accurate and reproducible; the position of the third limit cannot be determined accurately; it depends on experimental conditions.

54. Sorenson, S.C. and Savage, L.D., "Flammability Limits - A new Technique", Combustion and Flame, 24, 347 (1975).

A technique has been developed to study the extinguishment of premixed laminar flames. The technique utilizes a steady state burner with a tent flame surrounded by a separate stream of ignition gases, which are the produce gases of another premixed flame using the same fuel. Extinguishment is determined by observation of the shape of the tent flame as the composition of the test mixture of fuel-suppressant-air is changed so as to move toward a composition region of extinguishment. Results are reported for five methane-air flames and methane-suppressant-air flames containing the suppressants Helium, Argon, Nitrogen, carbon dioxide and Halon 1301 (CF3Br). The technique yields a slightly broader flammability region than other techniques previously used. The relation between the results of this technique and the standard flammability techniques is discussed.

55. Burgoyne, J.H. and Williams-Leir, G., "Inflammability of Liquids", Fuel, 28, 145 (1949).

"The conceptions of flash and fire points as applied to single liquids are examined and a number of values of the former characteristic calculated from standard inflammability data for the vapours. In general, values are slightly lower than those determined by standard methods. The inflammability in air of binary liquid mixtures of which one constituent is incombustible is examined experimentally in the particular case of n-hexane/difluorodichloromethane (freon 12) and a form of diagram is proposed to express the results. With the aid of a general diagram of the same kind the influence of mixture composition, temperature and volatility on the inflammability of binary mixtures of the same type is discussed."

56. Affens, W.A., "Flammability Properties of Hydrocarbon Solutions in Air", paper presented before Division of Petroleum Chemistry, American Chemistry Society, Chicago, Illinois, September 11-15 (1967).

Dr. Affens defines lower and upper flammability limits, flash points, flammability index, heats of combustion, stoichiometric concentration of vapor-air above liquids in this paper. Based on the principles of LeChatelier, Roault & Dalton, he develops mathematical expressions for predicting some of these flammable characteristics. The equation he develops shows how a relatively small amount of a contaminant that is highly volatile can make a relatively nonflammable fuel flammable.

57. Affens, W.A., "Flammability Properties of Hydrocarbon Fuels, Part 3 - Flammability Properties of Hydrocarbon Solutions in Air", NRL Report 6617, November 21, 1967.

"Review theoretical consideratives in support of observations concerning effects of admixing fuels of different volatility and flammability characteristics".

58. Zabetakis, M.S. and Richmond, J.K., "The Determination and Graphic Representation of the Limits of Flammability of Complex Hydrocarbon Fuels at Low Temperatures and Pressures".

"Flammability studies indicate that many precautions which are unimportant at room temperature and atmospheric pressure became important at low temperature and pressures." The authors describe some of the things that now must be given serious considerations. These include the selection of the test chamber, ignition source, and the handling of combustible-liquid test mixtures so as to prevent vaporization losses. The authors also describe the four categories of limit-of-flammability experiments. Results are given for a number of tests conducted on aviation and jet fuels.

59. Gerstein, M. and Steine, W.B., "Analytical Criteria for Flammability Limits".

"The one-dimensional laminar flame equations with singlestep Arrhenius kinetics, including both conduction and radiation heat loss to the surroundings, have been integrated. Adiabatic and nonadiabatic burning velocity, quenching distance, and an "apparent" flammability limit are calculated. It is shown that a fundamental flammability limit, independent of heat loss, does not exist even when radiation loss is included. The experimentally observed flammability limits are explained on the basis of the insensitivity of the critical fuel concentration for flame propagation to changes in tube diameter, when the radiation loss exceeds conduction loss. Flame thickness and inflection temperature are discussed as other possible criteria for "apparent" flammability limits. It is pointed out that safety criteria, based on flammability limits, should take into account the possible reduction of limits for very large systems.

Comparison is made with experimental values, in which pressure and diluent type and concentration are varied."

60. Macek, A., "Flammability Limits: Thermodynamics and Kinetics", Paper No. 75-28, presented at the 1975 Fall Meeting of the Western States Section of the Combustion Institute, October 1975.

"Extinction limits for both premixed and diffusion flames for n-alkanes and n-alcohols found in the literature are assembled. Several sets of theoretical flame temperatures corresponding to the limits are defined and presented. The implications of the view that flames fail to propagate at temperatures at which reaction rates become too low to overcome the dissipation processes are discussed. It is shown that at lean limits the excess oxygen does not act merly as a diluent but takes an active part in promoting the kinetics of flame reactions. The burning-rate data and the results of ignition experiments are shown to be pertinent to the interpretation of flammability limits. It is also shown that the assumption of thermodynamic equilibrium at the limits is unrealistic, so there is serious need for experimental temperature and concentration measurements in both premixed and diffusion flames. When the stipulation of thermodynamic equilibrium is removed, the chemical kinetic considerations suggest a simple qualitative explanation of the phenomenon of extinction by dilution."

61. Anson, D., "Influence of the Quality of Atomization on the Stability of Combustion of Liquid Fuel Sprays", Fuel, XXXII, 39-51 (1953).

Experiments were conducted on a small scale combustion rig burning kerosine sprayed from an air blast atomizer. By using a series of different injection pressures a family of curves was obtained representing the weak limits of stable combustion for each case. It was possible to relate the stability limit at a given air stream velocity to the spray mean particle size; in these experiments fine atomization was found to extend the range of stable combustion, but an optimum particle size may exist for any given scale of turbulence in the combustion chamber.

62. Bunev, V.A. and Babkin, V.S., "Deviations from the Le Chatelier Rule for the Limits of the Propagation of a Flame", Fizika Goreniya i Vzryva, 9, 605 (1973).

The paper is concerned with selective oxidation of one of the fuel components as one reason for deviations in Le Chatelier rule as used in evaluating explosive fuel mixtures. 63. Affens, W.A., Carhart, H.W. and McLaren, S., "Relationship Between Flammability Index and Temperature, and Flash Point of Liquid Hydrocarbon Fuels", presented before the Division of Petroleum Chemistry, American Chemical Society, April 1975.

"In earlier work a hydrogen flame ionization detector was used for measuring the flammability indices of hydrocarbon vapor-air mixtures. The response of the flame ionization detector was found to be proportional to flammability index for pure hydrocarbons and their mixtures (both vapors and liquids) in accordance with relationships derived from theoretical considerations.

The flammability indices of liquids were initially determined at a single temperature (51.7°C) and it was decided to extend the measurements to other temperatures in order to determine the relationship between flammability index and temperature of liquid hydrocarbons. Such information would be useful for extrapolating flammability indices from one temperature to another and for developing the relationship between flammability index and flash point."

### FLAME VELOCITY

64. Garner, F.H., Long, R., and Thorley, B., "Measurement of the Burning Velocities of Hydrocarbon-Air Mixtures Using a Nozzle Burner", Fuel, XXXIII, 394 (1954).

"A study has been made of the nozzle burner method of measuring burning velocities using benzene-air, propane-air and ethylene-air mixtures. Luminous, shadow and schlieren images of the flame cones have been measured. The luminous and schlieren images were found to be almost parallel over the major part of the nozzle flame cone. Whilst burning velocities deduced from the schlieren come by angle or frustum area methods very with the rate of flow of the unburnt gas, those obtained by the total area method are independent of the flow rate. However, when corrected for temperature, the burning velocities obtained by the latter method are low, as would be expected."

65. Andrews, G.E. and Bradley, D., "The Burning Velocity of Methane-Air Mixtures", Combustion & Flame, 19, 275 (1972).

"Results are presented for the variation of burning velocity with equivalence ratio for methane-air mixtures at one atmosphere pressure. Values were determined by the bomb-hot wire and corrected density ratio techniques, for combustion during the prepressure period. The former of these methods gives a maximum burning velocity of 45 ± 2 cm/sec, at an equivalence ratio of 1.07. Results are

compared with those of other workers and the reasons for discrepancies are discussed. The influence of pressure and unburnt gas temperature upon burning velocity are discussed also."

66. Andrews, G.E. and Bradley, D., "Determination of Burning Velocities: A Critical Review", Combustion & Flame, 18, 133 (1972).

"A critical survey is presented of the different experimental techniques for the measurement of burning velocity. Where possible, correction factors are derived to compensate for errors. The survey is carried out with particular reference to the maximum burning velocity of methane-air mixtures. Recommendations are made as to the most suitable methods of measuring burning velocity for both closed vessels and burners. The recommended value of the maximum burning velocity of methane-air is 45 ± 2 cm/sec at 1 atm and 298°K." (189 references)

67. Gerstein, M., Levine, O. and Wong, E.L., "Flame Propagation, II. The Determination of Fundamental Burning Velocities of Hydrocarbons by a Revised Tube Method", Journal American Chemical Society, 73, 418 (1951).

A method is described for the determination of fundamental rates of flame propagation in tubes. Data are presented for a variety of hydrocarbons including normal and branched alkanes, alkenes and alkynes as well as cyclohexane and benzene. The normal alkanes have a constant flame velocity except for methane which is slightly lower. Unsaturation increases the flame velocity in the order: alkanes < alkenes < alkynes. Branching reduces the flame velocity although the effect is small.

68. Flock, E.F., "Measurement of Burning Velocity", "High Speed Aerodynamics and Jet Propulsion, Vol. IX, Section K, Physical Measurements in Gas Dynamics and Combustion", Landenburg, R.W., Lewis, B., Rease, R.N. and Taylor, H.S., editors, Princeton University Press, Princeton, NJ (1954).

Subjects discussed are the effects of structure of the combustion wave, the burning velocities from stationary flames and from moving flames, the effect of the operating parameters on the burning velocity and a summary of recent (in 1954) measurements.

The operating parameter discussed in regard to burning velocity are fuel-oxygen ratio, fuel composition, mixture temperature, mixture pressure and additive and inert gases (42 references were cited).

69. Linnett, J.W., "Methods of Measuring Burning Velocities", Fourth Symposium (International) on Combustion, The Combustion Institute, 1952, pp. 20-35.

In this survey, methods of observation are discussed, followed by the methods used in measuring the burning velocities. These methods of measuring include: Egerton-Powley Flat Flame, Soap Bubble, Closed Spherical Vessel, Cylindrical Tube and Burner. A survey of the results from these various methods is presented. The author concludes that soap bubble method can be used to measure burning velocities where the gas can be enclosed satisfactory but cannot be used when slow flames are involved. The closed vessel method, at that time, had not been tested sufficiently. The tube method is useful with fast flames but not with slower flames. The burner method is very difficult to use and if one is not extremely careful, erroneous results are likely.

70. Polymeropoulos, E.C., "Flame Propagation in a Liquid Fuel Spray", Interim Report, Federal Aviation Administration, NAFEC, July 1972 to January 1974.

"The conditions for upstream flame propagation from a plane heat source were used for the calculation of the laminar burning velocity in a one-dimensional liquid fuel spray. Calculations were carried out for sprays where the burning was heterogeneously controlled through the transition to premixed gas behavior. The effect of fuel concentration, droplet size, and upstream temperature on the burning velocity were examined. Agreement was satisfactory between calculated results and existing experimental and theoretical data using monodisperse tetralin sprays. Experimental apparatus for the measurement of the burning velocity in a liquid fuel spray is described."

71. Polymeropoulos, C.E., "Ignition and Propagation Rates for Flames in a Fuel Mist", Interim Report No. FAA-NA-75-153, U.S. Department of Transportation, Federal Aviation Administration, July 1972-January 1975.

"A mathematical model was developed, which is capable of predicting the burning velocity in polydisperse airfuel sprays given the initial conditions of the liquid and gas phases. The analytical predictions were tested against previous experimental data, and the agreement was satisfactory.

The burning velocity in polydisperse kerosene-air sprays was measured at constant air-fuel ratio, and for various degrees of atomization of the spray in order to further check the predictions of the mathematical model. The results were also in good agreement with the analytical predictions."

72. Mizutani, Y. and Nishimoto, T., "Turbulent Flame Velocities in Premixed Sprays, Part I. Experimental Study", Combustion Science & Technology, 6, 1 (1972).

"This investigation was conducted to examine the influence of the flow velocity, turbulence intensity, droplet size distribution and fuel-air ratio on the turbulent flame velocities in premixed sprays. Premixed sprays of kerosene or light diesel oil were burnt either in a combustion chamber 450 mm long and of 77 mm x 75 mm cross section, or on an inverted-cone-flame burner of 21.6 mm diameter. The flame velocities were measured, and the physical structure of flames was observed by schlieren and high-speed photographies.

As a result, the following empirical relation was obtained

$$S_r = \frac{K}{d} (\phi - 0.012) (u')^{1.15} [m/sec]$$

where  $S_r$  denotes the turbulent flame velocity [m/sec], K is a constant whose value is 6,800 for kerosene sprays, d the Sauter mean diameter [microns],  $\phi$  the fuel-air ratio and u' the turbulence intensity of the approach flow. For light diesel oil sprays, the value of K was estimated to be 4,300 for u' around 0.1 m/sec from experiments on inverted-cone flames. It is noticed from the above relation that, besides the mixture strength, the average diameter of droplets, intensity of turbulence and volatility of the liquid fuel exert significant influences on the turbulent flame velocity."

73. Polymeropoulos, C.E. and Das, S., "The Effect of Droplet Size on the Burning Velocity of Kerosene-Air Sprays", Combustion and Flame, 25, 247 (1975).

The burning velocity of open inverted-cone shaped keroseneair sprays was measured at constant air-fuel ratio and for several degrees of atomization of the spray. The results show that as the degree of atomization in the spray increases, the burning velocity first increases to a maximum value, and then decreases to the burning velocity approaching that of a premixed gas flow.

74. Hayashi, S., Kumagai, S., and Sakai, T., "Propagation Velocity and Structure of Flames in Droplet-Vapor-Air Mixtures".

"Flame speeds of ethanol and n-octane mixtures of monosized droplets, vapor and air have been measured to study the fundamental aspects of spray combustion. The state of the mixtures, which were prepared in an apparatus similar to the Wilson cloud chamber, was experimentally verified in detail. A rugged, undulated and thickened flame front with the cellular structure is peculiar to the flame propagation in the mixtures containing large droplets and is in sharp contrast to the smooth and continuous flame

front observed with premixed gas mixtures or mixtures containing droplets of much smaller size. This difference in the flame structure, caused by the heterogeneous nature of the unburned mixture, can explain the observation that the burning velocity in a droplet-vapor-air mixture is larger than in a homogeneous mixture of the same overall fuel-air ratio, even on the lean side of the stoichiometric point, provided that the droplet diameter is large enough. Another aspect of the flame propagation in the droplet-vapor-air mixtures can be interrupted in terms of the effective fuel-air ratio, which accounts for the incomplete evaporation and mixing of the liquid fuel."

#### APPENDIX B

# Light Scattering Method of Droplet Size Measurement

In principle, when a spherical object is placed in a beam of collimated light, the light is diffracted around the object such that the shadow of the object on a screen will have a bright or dark spot on its center depending on the distance between the object and the screen. This type of diffraction was discovered by Fresnel. Fraunhofer diffraction is obtained by passing this diffracted light through a condensing lens onto a screen placed at the focal point of the lens (see Figure 1). The diffraction pattern caused by a single object will be a series of concentric light and dark bands of decreasing intensity. Mathematically, the diffraction pattern is described in terms of Bessel functions  $J_1$  ( $\alpha$ ); the argument  $\alpha$  is

$$\frac{\pi \Theta d}{\lambda}$$
 or  $\frac{\pi r d}{\lambda F}$ 

where:

0 = angle of scattered light relative to the axis of the collimated beam

d = diameter of the object

r = spacial displacement of the light intensity away from
the central spot

F = the focal length of the lens,

 $\lambda$  = wavelength of the light.

It is obvious that for a given goemetry, the distribution of light intensity depends only on the particle size and that smaller particles scatter at larger angles. With certain restrictions to avoid multiple scattering, the scattered light from a large number of spheres of the same radius will superimpose and the intensities are additive. If on the other hand, there is a distribution of sizes among the spheres, then the intensities will still be additive but the pattern will smear and give a continuous distribution of light rather than concentric bands. The intensity pattern for a polydisperse spray is, therefore,

$$I(r) = \sum_{i=1}^{N} \frac{n_i \pi^2 d_i^4}{16\lambda^2 F^2} \left[ \frac{2 J_1 \frac{\pi d_i r}{\lambda F}}{\frac{\pi d_i r}{\lambda F}} \right]^2$$

where  $\mathbf{n}_i$  is the number of particles of size  $\mathbf{r}_i$  and N is the total number of sizes.

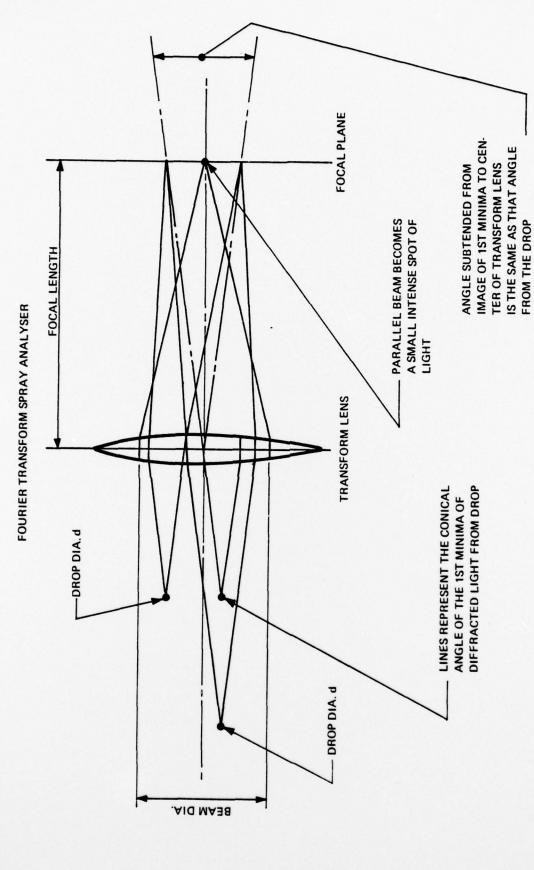


FIGURE 1. EFFECT OF DROP POSITION ON DIFFRACTION PATTERN

# Experimental Considerations

The scattering pattern we have just described is in actuality only an approximation of the total solution. The Fraunhofer approximation places certain restrictions on particle size, wavelength, and measurement distance on the droplet size. In particular, for circular scatterers (or spherelets, if the distance between the spherelet and measurement plane is large compared to the diameter) the Fraunhofer approximation is valid if

$$z \gg \frac{\pi d^2}{\lambda}$$

where:

z = distance from scattering volume to the measurement plane

d = diameter of scatterer

 $\lambda$ = wavelength of illuminating radiation

For red light from a helium-neon laser ( $\lambda$  = 0.633 µm) and droplets in the 10 µm range,

$$z >> \frac{3.14 (10 \mu m)^2}{0.633 \mu m} = 496 \mu m$$

or

a condition easily met in the intended application where z = 305 mm.

With the Fraunhofer (or far-field) approximation, the first dark ring in the diffraction pattern generated by a thin volume of scatterers of a uniform diameter occurs when

$$J_1(\alpha) = 0 \text{ at } \alpha = 3.83$$

Since the first zero is of primary interest in determining the diameter of the scatterer, the above may be reduced to simply

$$r = 3.83 \frac{\lambda F}{\pi d}$$

Or, in terms of a scattering angle,

$$\Theta = \tan^{-1}\left(\frac{1.22\lambda}{d}\right)$$

In the intended application where droplet diameters range from 1  $\mu\text{m}$  to 100  $\mu\text{m},$  the expected solid scattering angles for these respective droplet diameters are

$$\Theta_1 = \tan^{-1} \left( \frac{(1.22) (0.633)}{(100)} \right) = 0.44^{\circ} \text{ for } 100 \text{ } \mu\text{m } \text{ diameter}$$

and

$$\Theta_2 = \tan^{-1}\left(\frac{(1.22) (0.633)}{(1)}\right) = 37.7^{\circ} \text{ for 1 } \mu\text{m diameter.}$$

Although the difference in the two scattering angles is large and should present no difficulties in measurement, care must be taken in performing the experiments to ensure accurate results. One source of possible error is multiple scattering, which can occur if the thickness of the scattering volume is not kept as small as possible. This suggests the use of a fan shaped or collimated spray of fuel. A check which can be performed to detect the presence of multiple scattering is to measure signal intensity as a function of illuminated area. If the signal at a given angle is less than proportional to illuminated area, then multiple scattering is taking place and the scattering volume is too thick.

Additionally, because the fuel droplets will have a range of diameters, the diffraction pattern will be a smear, as opposed to the discrete set of concentric circles described by the Bessel function. For example, the presence of 1  $\mu m$  range scatterers (mist) in the company of 100  $\mu m$  range scatterers (spray) should produce a very noticeable increase in the high spatial frequency content of the diffraction pattern. One method which may be employed merely to detect the presence of scatterers smaller than the range encompassed by the host droplets is to place a detector at an angle to the scattering volume such that the light component scattered to it from the host droplets is negligible compared to the contribution expected from the smaller droplets. In this application, this angle would be in the neighborhood of 25°.

Most essential to determining the mean size distribution of smaller droplets is accurate measurement of the diffraction pattern. The accuracy depends upon how well the diffraction intensity can be measured at specified displacements from the center-line (primary laser beam). In the present experiments, the position of the detector is controlled by a micrometer driven optical mount in which a dial indicator was installed to make exact displacement readings.

The solution to the problem of determining the size distribution from measured scattering intensities has been programmed by Watkins and Opdyke of the Combustor Section of AVCO/Lycoming, a gas turbine engine company. The program breaks the distribution up into size groups which can be fitted to a Rosen-Ramler distribution function for sprays. The program was written for the Hewlett-Packard 9820 calculator, a facility of AFLRL. In practice, relative intensities are measured at specified scattering angles pre-defined in terms of the size groups and the manner in which the program was written.